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(54) Title: NOVEL DELIVERY SYSTEM FOR INORGANIC SUNSCREENS

(57) Abstract: A novel delivery system for physical, inorganic sunscreens for use in sunscreen preparations is disclosed. One or more inorganic suscreen agents, such as micronized zinc oxide and micronized titanium dioxide, is dispersed in a solid dispersion vehicle preferably selected from the group consisting of stearyl benzoate, behenyl benzoate and arachidyl benzoate, to thereby produce a solid dispersion of inorganic sunscreen agents for use in sunscreen preparations. The micronized sunscreens are embedded in a solid product which does not require mixing or regrinding before use. A solid formulation avoids the problems inherent in a fluid emulsion or dispersion, i.e., migration or settling out of the contents, which requires mixing and/or regrinding before use.

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

10

TITLE: NOVEL DELIVERY SYSTEM FOR INORGANIC
SUNSCREENS

15

SPECIFICATION

BACKGROUND OF THE INVENTION

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1. FIELD OF THE INVENTION

The present invention relates to a novel delivery system for physical, i.e., inorganic, sunscreens, and the use of said delivery system in the preparation of stable sunscreen compositions.

25

2. Description of the Related Art

Sunscreen products may be emulsions, creams, lotions, gels, liquids, solid sticks, aerosols and all other forms of cosmetic compositions. Sunscreen compositions are applied topically to human skin to protect the skin against UV radiation damage. The term "sunscreen" herein is meant to include tanning lotions, sunscreens and sunblockers intended for topical application to human skin and/or hair to protect against ultraviolet radiation from the sun.

35

Active sunscreen ingredients which filter UV-A or UV-B rays harmful to the skin may be organic sunscreens such as PABAs (p-aminobenzoic acids), benzophenones, salicylate esters, and di-oxybenzone, octyl methoxycinnamate, and mixtures thereof, or physical sunscreens, which are inorganic compounds such as titanium dioxide and zinc oxide.

40

5 It has been found that inorganic sunscreen agents such as
TiO₂ and zinc oxide are superior to customary organic
sunscreen agents, as they are less irritating and offer
better or broader UV protection. However, inorganic
10 sunscreen agents are difficult to incorporate into sunscreen
formulations. The invention relates to a novel delivery
system for these inorganic sunscreen agents.

15 Micronized, also referred to as "microfine", physical sun-
screens, such as zinc oxide and titanium dioxide, have a
preferred particle size less than 1 micron. These particles
of less than 1 micron or submicron size are effective sun-
block agents. They do not scatter light, and therefore are
useful in cosmetics, make-up, and sunscreen applications.
20 Particles larger than 1 micron are undesirable as they inher-
ently result in so-called "whitening effects".

25 The problem is that micronized TiO₂ and zinc oxide parti-
cles are susceptible to agglomeration after their production,
during the post-manufacture period of transportation and
handling. This tendency to agglomerate into clumps of much
30 higher particle sizes (greater than 1 micron) reduces the
efficacy of TiO₂ and Zinc Oxide as UV sunscreens and in-
creases their white appearance on the skin. The ability of
the micronized, inorganic sunscreens to function as UV-ray
blocking agents is impeded if the micronized particle size is
35 enlarged by agglomeration or dusting. Agglomeration also
results in the deterioration of emulsions including such
particles and negatively affects their stability and shelf-
life during storage.

Not only are the fine, micronized inorganic sunscreen
powders susceptible to agglomeration, but these particulates
also have a tendency to show dusting effects during handling,
such as charging to the mixing vessels.

5 The disadvantages of dusting and agglomeration, or re-agglomeration, are avoided by the novel delivery system of this invention.

10 Numerous references describe the use of micronized zinc oxide and titanium dioxide as sunscreen materials, in various formulations. None of these references, however, teach or suggest the specific novel delivery system of physical sunscreens of this invention, which prevents inorganic sunscreen pigment particles from accumulating to give agglomerates.

15 U.S. Patent No. 4,323,693 to Scala, Jr. discloses a benzoic acid ester of isostearyl (C18) alcohol.

20 U.S. Patent No. 4,917,882 to Stobridge discloses a gel-type sunscreen composition and a method of making same, comprising combining a sunscreen agent, polyethylene, and a benzoic ester, agitating and heating the mixture to a temperature and for a time sufficient to dissolve the polyethylene in the benzoate ester, and after the polyethylene is dissolved in the benzoate ester, cooling the mixture while agitating to produce a gelled sunscreen composition.

25 U.S. Patent No. 5,340,567 to Cole et al. discloses a sunscreen composition comprising an extending medium such as a carrier or vehicle (such as mineral oil and polyethylene) and a synergistic combination of microfine titanium dioxide having a particle size of less than about 35 μ and zinc oxide having a particle size of less than about 50 μ . The particle sizes of the titanium oxide and zinc oxide are critical for obtaining a composition which is invisible on the skin.

5 U.S. Patent No. 5,417,961 to Nearn et al. discloses a sun-
screen composition comprising a water-in-oil emulsion which
comprises an aqueous phase and an oil phase, the oil phase
comprising polyethylene and an organic sunscreen agent.
Microfine zinc oxide having a particle size in the range of
10 from about 0.01 microns to about 0.25 microns is suspended in
the oil phase.

U.S. Patent No. 5,468,471 to Zecchino et al. discloses an
organic dispersion of microfine titanium dioxide, of a parti-
15 cle size prior to agglomeration of about 10nm to about 100nm,
and a cosmetically acceptable branched chained organic com-
pound, preferably octyldodecyl neopentanoate, without any
dispersing agent. The dispersion is prepared by subjecting
the microfine titanium dioxide and the branched chain organic
20 compound to a ball mill, roller mill or ultrasonic mixer, to
grind the titanium dioxide in the organic compound and to
disperse the titanium dioxide in the organic compound.

U.S. Patent No. 5,476,643 to Fogel discloses use of two
25 neopentyl glycol diesters as wetting, dispersing, spreading
and detergent agents for micronized titanium dioxide and zinc
oxide.

U.S. Patent No. 5,498,406 to Nearn et al. discloses a sun-
30 screen composition in oil-in-water emulsion form having about
0.5% to about 5% by weight of microfine titanium dioxide
having a particle size of less than about 100 nm uniformly
suspended therefrom, the composition further comprising a
dispersing agent comprising a long chain saturated primary
35 alcohol having an average of from about 25 to about 45 carbon
atoms in the long chain, to stabilize the emulsion.

U.S. Patent No. 5,543,136 to Aldous discloses making and
using a water-in-oil emulsion comprising zinc oxide and an

5 agent selected from the group consisting of tridecyl
neopentanoate, C₁₂₋₁₅ alkyl benzoate, octyl neopentanoate and
mixtures thereof in the emulsion's oil phase; titanium diox-
ide in the emulsion's water phase, a sunblocking agent, and
oil phase emulsion components.

10

U.S. Patent No. 5,605,652 to Tapley discloses a method of
preparing sunscreens in which a dispersion of zinc oxide
particles in an oil is formed by milling in the presence of a
particulate grinding medium and mixed with cosmetically
15 acceptable materials. A mixed oxide dispersion comprising an
oil, particles of zinc oxide, particles of titanium oxide,
and an organic dispersing agent is formed.

20

U.S. Patent No. 5,573,753 to Tapley discloses a method of
preparing sunscreen containing a zinc oxide dispersion com-
prising milling a particulate zinc oxide in an oil in the
presence of a particulate grinding medium and an organic
dispersing agent.

25

U.S. Patent No. 5,599,529 to Cowie discloses an oil
dispersion comprising an oil, titanium dioxide particles, and
an organic dispersing agent for the particles. The disper-
sion has a solids content of greater than 40% by weight. The
oil is selected from the class of oils consisting of fatty
30 acid esters, fatty alcohols and saturated fatty acid di-
esters. The oil dispersion is prepared by milling particu-
late titanium dioxide in an oil in the presence of a particu-
late grinding medium and an organic dispersing agent.

35

U.S. Patent No. 5,725,844 to Gers-Berlag et al. discloses
a sunscreen in the form of an oil in water emulsion or a
hydro-dispersion, comprising one or more silanized hydropho-
bic inorganic pigments incorporated into the oily phase of
the emulsions or hydrodispersions, one or more oil-soluble UV

5 filter substances, one or more film-forming agents and optionally one or more water-soluble UV filter substances.

10 U.S. Patent No. 5,730,993 to Allard et al. discloses a sunscreen comprising an ultrafine oil-in-water emulsion resulting from phase inversion of homogeneously and finely dispersed particulate of at least one nanopigment comprising a metal oxide, such as titanium dioxide or zinc oxide, having a particle size from 100 nm to 1,000 nm.

15 U.S. Patent No. 5,747,012 to Dahms discloses a process for preparing a sunscreen comprising mixing a dispersion in an oil of particles of metallic oxide having an average particle size of less than 0.2 micrometers, with one or more emulsifiers, an aqueous phase, and a hydrophilic organic sunscreen,
20 under conditions in which an emulsion is formed. The emulsion may be an oil-in-water emulsion or a water-in-oil emulsion. The dispersion of particles of metallic oxide is prepared by milling the particulate metallic oxide in the oil in the presence of a particulate grinding medium and in the
25 presence of a dispersing agent.

U.S. Patent No. 5,776,440 to Forestier et al. discloses a sunscreen composition comprising at least one coated nanopigment of metallic oxides, and at least one UV-screening
30 fat-soluble polymer, in a cosmetically acceptable carrier.

U.S. Patent No. 5,788,952 to Gers-Berlag et al. discloses a sunscreen comprising inorganic micropigments and optionally additional organic UV filter substances. The formulation is
35 a hydrodispersion consisting of an inner lipid and an outer aqueous phase, free of emulsifiers, with the organic micropigments incorporated in the preferably liquid lipid phase of the hydrodispersion.

5 U.S. Patent No. 5,817,298 to Galley et al. discloses a
sunscreen composition comprising a water-in-oil emulsion
which comprises 0.5 to 50% by weight of titanium dioxide
particles having a mean particle size of less than 100 nm,
each of the particles being substantially coated with
10 phospholipid, together with a cosmetically acceptable car-
rier. Coating titanium dioxide particles with phospholipid
reduces their tendency to clump.

The references describe various methods to prevent or
15 reduce clumping of micronized physical (inorganic)
sunscreens, so as to improve the efficacy of the sunscreen
composition. However, among the foregoing patents, none
disclose the specific delivery system for physical (inorgan-
ic) sunscreens of the invention or the use of such systems in
20 the production of stable sunscreen compositions.

Thus, none of the references appear to teach or suggest a
delivery system for physical (inorganic) sunscreens in which
micronized sunscreens are embedded in a solid product which
25 does not require mixing or regrinding before use. A solid
formulation avoids the problems inherent in a fluid emulsion
or dispersion, i.e., migration or settling out of the con-
tents, which requires mixing and/or regrinding before use.

30 OBJECTS AND SUMMARY OF THE INVENTION

It is an object of the present invention to provide a
novel delivery system for physical, inorganic sunscreens
which results in non-agglomeration of the fine, micronized
powders.

35 It is another object of the invention to provide a novel
delivery system for physical, inorganic sunscreens for use in
sunscreen applications.

5 It is yet another object of the invention to a novel delivery system for physical, inorganic sunscreens which results in non-dusting of the resulting fine, micronized powders.

10 It is a further object of the invention to provide a novel delivery system for physical, inorganic sunscreens which results in micronized powders which are easily re-meltable.

15 Yet another object of the invention is to provide a novel delivery system for physical, inorganic sunscreens which results in fine, micronized particles which are easily dispersible in the commonly used systems for sunscreen applications.

20 Further, it is an object of the invention to provide a method for dispersing micronized titanium dioxide and zinc oxide particles prior to utilization in cosmetic sunscreens.

25 Another object of the invention is to provide a novel delivery system for physical, inorganic sunscreens which results in micronized powders which are easily admixable in the oil phases of cosmetic creams, lotions, etc.

30 It is another object of the invention to provide a novel delivery system for physical, inorganic sunscreens which results in micronized powders which are easily grindable, if necessary.

35 Yet another object of the invention is to provide a novel delivery system for physical, inorganic sunscreens which results in more effective sunscreens.

 It is another object of the invention to provide a novel delivery system for physical, inorganic sunscreens which

5 results in sunscreens which are more effective over a longer period of time, reducing the need to reapply the sunscreen as frequently as would otherwise be necessary.

10 It is another object of the invention to provide a novel delivery system for physical, inorganic sunscreens which results in increased stability and shelf-life of the physical sunscreens and/or of the sunscreen preparations.

15 These and other objects are accomplished by providing an improved delivery system for micronized inorganic sunscreens in which micronized inorganic sunscreens are embedded in a solid product which does not require mixing or regrinding before use. A solid formulation avoids the problems inherent in a fluid emulsion or dispersion, i.e., migration or settling out of the contents, which requires mixing and/or
20 regrinding before use. The micronized inorganic sunscreen is mixed into a pre-melted, solid vehicle such as Stearyl Benzoate (FINSOLV® 116 from Finetex, Inc., Elmwood Park, N.J.) and Behenyl Benzoate (FINSOLV® 137 from Finetex, Inc.,
25 Elmwood Park, N.J.), the mixture is mixed for intimate blending, and the solid dispersion discharged and cooled on flaking equipment or on any cooled surface.

30 The disadvantages of the known delivery systems for physical, inorganic sunscreens are overcome by the process of the present invention. The micronized physical sunscreens of the invention are non-agglomerating and non-dusting, and thus retain their effectiveness in sunscreen applications.

35 DETAILED DESCRIPTION OF THE INVENTION

This is accomplished by providing a novel delivery systems for physical, inorganic sunscreens. The micronized zinc oxide or titanium dioxide is mixed into a pre-melted, solid

5 dispersion vehicle. The melting is done above the melting
point of the respective vehicle. The mixture is subjected to
mixing by means of a mixer for intimate blending. The mix-
ture is then discharged and cooled on flaking equipment or on
10 any cooled surface, especially cooled, flat, stainless steel
belts. The product so cooled is a solid material, preferably
in the form of flakes.

Titanium dioxide is an inorganic pigment widely used in
paints, and in cosmetic products such as bar soaps, to en-
15 hance whiteness. The particle size of commonly used titanium
dioxide is generally between 150 and 350 mu. Titanium
dioxide also absorbs and scatters UV-radiation.

Zinc oxide is an inorganic substance which finds use as a
20 white pigment in paints, papers and polymers, and as a physi-
cal sunblock by scattering and absorbing ultraviolet radia-
tion.

Naturally, the grade of titanium dioxide and zinc oxide
25 used as sunscreen agents in cosmetic preparations is differ-
ent than that the grade used as a pigment in manufacturing
paint, paper or plastics. The particles of inorganic
sunscreen to be used in the present invention are fine,
micronized particle size. If the particles are too large,
30 over a micron, they are not effective as sunscreens. The
particles may optionally be surface-treated with organic or
inorganic coatings, such as aluminum stearate, or one or more
oxides or hydrous oxides of aluminum, silicone, titanium,
magnesium or zinc, etc., to prevent graying in sunlight.

35

Thus, inorganic sunscreens such as titanium dioxide or
zinc oxide having a particle size less than 1 micron is used
in the invention. Preferably, inorganic sunscreens having a

5 particle size between 0.1 and 0.50 micron, and most preferably from about 10 nm to about 100 nm, are used.

10 Inorganic sunscreen agents such as titanium dioxide and zinc oxide have a tendency to agglomerate that reduces their efficacy, results in an unpleasant feel and gives a white appearance on the skin.

15 In general, the preferred method of producing the delivery system of the invention begins by melting the solid vehicle, and combining between about 10% and 60% of inorganic
20 sunscreen agent and between about 40% and 90% of said melted vehicle, such as benzoate ester, depending upon the type of sunscreen preparation and its desired sunscreen efficacy or SPF. This mixture is then agitated and heated to a temperature and for a time sufficient to disperse the sunscreen
25 agent in the vehicle. Any mixer or stirrer is suitable. Preferably, the temperature to which the mixture is heated will be above about 100°C. After the inorganic sunscreen is all dispersed in the vehicle, the mixture is cooled while
30 agitating with a mixer or stirrer to thereby form a solid dispersion of the physical sunscreen. Preferably, the temperature to which the mixture is cooled while agitating is below about 60°C. The mixture is preferably homogenized by passing it through a homogenizer or other means, such as an in-line mill.

35 The solid dispersion vehicle is preferably Stearyl Benzoate (FINSOLV® 116 from Finetex Inc. of Elmwood Park, New Jersey, U.S. Patent No. 4,323,694), Behenyl Benzoate (FINSOLV® 137 from Finetex Inc.), or Arachidyl Benzoate (a C-20 benzoate). Other dispersion vehicles which may be used in the process of the invention are solid fatty alcohols such as:

5 Cetyl Alcohol
 Stearyl Alcohol
 Cetearyl Alcohol
10 Behenyl Alcohol
 Arachidyl Alcohol or
15 Higher Alcohols (C22+...)

The foregoing list is only exemplary of the type of esters on which the delivery system may be based, and, as such, is not to be considered limiting.

20

Additives which offer additional dispersing effects to the sunscreen compositions may be added in the vehicle delivery system at the time of melting of the dispersion vehicle. These are liquid emollients which may be added to the system in small quantities and do not affect the solid characteristics of the vehicle delivery system. Among them are:

25

C₁₂₋₁₅ Alkyl Benzoate (FINSOLV® TN from Finetex Inc., U.S. Patent No. 4,323,694)

30

Octyl Dodecyl Benzoate (FINSOLV® BOD from Finetex Inc.)
PPG-15 Stearyl Ether Benzoate (FINSOLV® P from Finetex Inc., U.S. Patent No. 4,791,097)

Dipropylene Glycol Dibenzoate (FINSOLV® PG-22 from Finetex Inc.)

35

The above and similar compounds are liquid emollients which promote the dispersion of the particulate inorganic sunscreen in the dispersion vehicle. Their addition is preferred but is not required. The quantity of the dispersing agent used depends on various factors but generally an amount from 1% to 20%, preferably from 1% to 10% by weight based on the weight of the particulate matter may be added.

40

5 These benzoate additives offer a processing benefit for
the use of the solid vehicle delivery of micronized zinc
oxide and titanium dioxide. Benzoate esters offer dispersing
effects to the sunscreen compositions. The liquid benzoate
esters identified above offer particularly effective dispers-
10 ing effects and emolliency in the final compositions.

 The micronized physical sunscreens of this invention in
the solid dispersed form as prepared by the invention process
may be incorporated into various cosmetic and personal care
15 products such as hand and body creams, a suspension or dis-
persion in solvents or fatty substances, or alternatively, as
lotions, ointments, oils, gels, emulsions such as a cream or
a milk, solid sticks, facial cosmetics, lip balm products and
the like, and may optionally be packaged as an aerosol or in
20 the form of a foam or a spray.

 The amount used in such compositions is dependent on the
type of composition, the type and quantity of other ingredi-
ents, such as cosmetic ingredients used, and the amount and
25 type of functional additives that are utilized. Typically,
the percent concentration of the micronized zinc oxide or
titanium dioxide used may vary from 0.5% to 65%, by weight,
depending upon the intended use of the particular sunscreen
composition. For instance, low amounts are required in
30 suntanning products which are not intended to prevent the
sun's rays from reaching the skin. More substantial amounts
are required for sunblocks products which are intended to
prevent substantially all of the sun's rays from contacting
the skin. A preferred, usable form carries about 60 parts of
35 micronized zinc oxide or titanium dioxide per 40 parts of the
benzoate ester or other dispersion vehicle.

 The delivery system of the invention is advantageous as it
results in micronized physical sunscreens which are:

- 5 - non-dusting
- non-agglomerating
- easily re-meltable
- easily dispersible in the commonly used systems for
 sunscreen applications
- 10 - easily admixable in the oil phases of the cosmetic
 creams, lotions, etc.; and
- easily grindable, if necessary.

15 Another advantage is that the delivery system of the invention allows for pre-mixed, pre-dispersed sunscreen compositions suitable for long-term storage as stable, solid dispersions. Such compositions preferably comprise 10% to 60% by weight of inorganic sunscreen agents in a solid dispersion.

20 Another advantage is the solid dispersion has excellent homogeneity, i.e., the inorganic sunscreen pigments are very well dispersed in the solid dispersion, and when mixed into a sunscreen preparation for topical application.

25 Thus, the present invention provides novel stable and homogeneous sunscreen compositions comprising inorganic sunscreen pigments in a cosmetically acceptable vehicle of the solid dispersant type.

30 The delivery system of the invention makes the zinc oxide or titanium dioxide particles more dispersible, so that higher concentrations than were possible before may now be incorporated into stable fluid emulsions, dispersions, creams, lotions, ointments, etc. Thus, the delivery system
35 of the invention enhances the dispersibility of the inorganic sunscreen particles, contributing to an improvement in the UV sunscreens efficiency and long-term stability of sunscreen compositions incorporating same.

5 Once the solid dispersion of zinc oxide or titanium dioxide
in the dispersion vehicle, preferably a solid benzoate ester,
has been prepared, it is ready for use in cosmetic sunscreen
preparations as the active sunscreen ingredient.

10 In sum, the delivery system of the invention provides a
means for delivering micronized inorganic sunscreens embedded
in a Finsolv® benzoate ester product. There are currently no
other solid dispersions of physical sunscreens on the market
of the type of this invention. Liquid dispersions have
15 drawbacks as the micronized sunscreen settles out over time,
and must be stirred vigorously before use. In contrast, the
invention contemplates a homogeneous, solid material into
which the micronized physical sunscreen has been dispersed,
which does not require mixing or regrinding prior to use.

20

 The solid dispersions of physical sunscreens so prepared
provide a convenient form of sunscreen which may be mixed
with one or more cosmetically acceptable materials in the
customary manner to formulate various types of sunscreen
25 compositions. These may be used for cosmetic and/or
dermatologic protection from the sun, and for the treatment,
care and cleansing of the skin or hair, and as make-up. The
formulation may take the form of anhydrous gel-type sunscreen
compositions, oil in water emulsions and water in oil emul-
30 sions. There are no limitations on the use of the solid
dispersions of the invention in sunscreen compositions. The
cosmetic and dermatologic preparations are used by applying
them to the skin and/or the hair in effective amounts in the
customary manner.

35

 Preferred compositions include emollients, selected from
the group consisting of lanolin, isopropyl myristate, glycer-
yl stearate, cetyl alcohol, and dimethicone, and combina-

5 tions thereof. The compositions may include further ingredi-
ents such as customarily used in such preparations, i.e.,
conventional adjuvants, including organic or inorganic
10 sunscreens, fragrances, preservatives, bactericides, emulsi-
fying agents, stabilizers, dispersants, anti-oxidants, thick-
eners, moisturizers, moisture-retaining agents, fats, oils,
waxes, foam stabilizers, electrolytes, water-proofing agents,
15 fillers, humectants, corrosion inhibitors, solubilizing
agents, coloring pigments, vitamins, deodorizing agents,
anti-perspirants, insect repellents, alkalizing or acidify-
ing agents, shaping agents, propellants, or any other ingre-
20 dient generally used in the cosmetic field, as is known in
the art.

Typically, in a general method for preparing a sunscreen
20 lotion, hydroxyethyl cellulose (Natrosol® from Hercules, Inc.
of Wilmington, DE) is dispersed in water and heated to 75°C
to form a first Phase A. A second Phase B is prepared by
separately heating glyceryl monostearate, DEA Oleth-3 phos-
25 phate, and cyclomethicone (345) to 75°C, and then adding
Phase B to Phase A with mixing. Phase C, comprising the
solid dispersion of inorganic sunscreen prepared as described
above, is added in small portions with vigorous stirring by
means of a mixer over a period of one hour. The mixture is
30 cooled with stirring to 40°C. The loss of water is adjusted.
When the temperature reaches 40°C, Phase D comprising
propyleneglycol, diazolidinylurea, methyl paraben and propyl
paraben (Germaben II from ISP, Wayne, NJ) is added. The
lotion is then homogenized with a hand homogenizer.

35 The ingredients and quantities used in any particular sun-
screen composition will depend on the type of sunscreen, and
the degree of SPF desired.

5 A further advantage of the delivery system of the invention is that lotions prepared thereby are invisible, i.e., do not have a whitening effect on the skin.

10 Another advantage is that sunscreen compositions made using the method of the invention have a pleasing appearance, because the benzoate ester provides a generally translucent medium. As a result, the composition can include dyes and the like to confer desirable colors to the composition.

15 Yet another advantage is that the compositions have been found to possess high substantivity, i.e., retention on the skin, even when reasonably immersed in water for 30 - 60 minutes or more.

20 A further advantage of the invention is that the consistency, i.e., the viscosity and homogeneity, of the composition is stable over a wide range of temperatures. The preferred composition of the invention is generally stable at temperatures below 0°C and above about 55°C. It will not
25 liquify in high temperatures or freeze in low temperatures.

 The delivery system of the invention provides a convenient and economical method for producing a wide variety of sunscreens.

30 The following are non-limiting examples of the novel delivery system for physical, inorganic, sunscreens, and the use of said delivery system in the preparation of stable sunscreen compositions. For ease of identification, each
35 example is identified by both an Example Number and a Reference No., where applicable. Specifically, Example Nos. 1 through 11 are processes for pre-incorporation of physical sunscreens into benzoate esters according to the invention process. Example Nos. 12-14, 16, 18, 20, 22, and 24 are

5 comparative examples showing processes for preparing
sunscreen compositions using individual separate components,
i.e., benzoate esters and physical sunscreens are added
separately to make the lotions, as is known in the art.
Examples Nos. 15, 17, 19, 21, 23, and 25 are examples of
10 processes for preparing sunscreen lotions using the products
of this invention which are blends of physical sunscreens and
benzoate esters, as prepared in Example Nos. 1 through 11.

15 In the Examples, as well as throughout this application,
the chemical and scientific symbols have their customary
meanings and all percents are weight percents unless other-
wise specified.

20 By "other process" in the Examples below, unless otherwise
specified, is meant a process of the prior art as described,
wherein benzoate esters and physical sunscreens are added
separately to make the sunscreen lotions.

25 The following examples are intended to illustrate the scope
of the present invention. As these examples are illustrative
only, the invention should not be inferred to be limited to
these examples.

30

35

5

TABLE I

	TRADE NAME/ PRODUCT	INCI NAME	SOURCE
10	FINSOLV® TN*	C12-15 Alkyl Benzoate	Finetex® Inc.,
	FINSOLV® TPP**	C12-15 Alkyl Benzoate/ Dipropylene Glycol Dibenzoate/PPG-15 Stearyl Ether Benzoate	Finetex® Inc.,
15			
	FINSOLV® 116	Stearyl Benzoate	Finetex® Inc.,
20	FINSOLV® 137	Behenyl Benzoate	Finetex® Inc.,
	NATROSOL	Hydroxyethyl Cellulose	Hercules, Inc., Wilmington, DE
25	Z-COTE	Zinc Oxide	Sun Smart, Inc., Wainscott, NY
	Z-COTE HP1	Zinc Oxide/dimethicone	Sun Smart, Inc.,
30	T-COTE 031	Titanium Dioxide/Dimethicone	Sun Smart, Inc.,
	UV TITAN (X-161)	Titanium Dioxide/Stearic Acid/Alumina	Presperse, Inc., Piscataway, NJ
35	UV TITAN (M-262)	Titanium Dioxide/ Dimethicone/Alumina	Presperse, Inc.,

* Patent Pending

40 ** Patent No. 4,791,097

5 EXAMPLE #1 (121.84)

DISPERSION OF TITANIUM DIOXIDE (T-COTE 031 FROM SUNSMART,
INC., WAINSCOTT, NY) IN FINSOLV® 137 & FINSOLV® TPP

10 In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 22.5 grams FINSOLV® 137 and 10 parts (5.0
grams) FINSOLV® TPP. Heated the mixture to 60°C to attain
clear liquid on hot water bath. Started adding titanium
dioxide powder (22.5 grams) in five equal portions over a
period of two hours with good mixing and raising the tempera-
15 ture to 65°C and maintaining the fluidity of the mixture all
the time. Raised the temperature to 70°C and held for one
hour with good mixing with a stirrer. Cooled the mixture to
60°C and discharged onto glass pyrex plate to obtain the
flaked form.

20 Remarks:

- 1) Addition of titanium dioxide powder into liquid ester did
not make any globules or clumps.
- 2) The mixture was in liquid form throughout the process.
- 25 3) Flakes were not too hard.
- 4) After discharge onto plate, there was no powder on the
surface of the flakes.

30 Example #2 (121-85)

DISPERSION OF ZINC OXIDE (Z-COTE) IN FINSOLV® 116

35 In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 25 grams FINSOLV® 116. Melted the flakes at
60°C to attain clear liquid on hot water bath. Started
adding zinc oxide powder (25 grams) in five equal portions
over a period of two hours with good mixing and raising the
temperature to 65°C and maintaining the fluidity of the
mixture all the time. Raised the temperature to 70°C and
40 held for one hour with good mixing. Cooled the mixture to

- 5 60°C and discharged onto glass pyrex plate. Remarks:
- 1) Addition of zinc oxide powder into liquid ester did not
make any lumps, globules or agglomerates.
- 2) The mixture was liquid all the time during the addition
10 of powder.
- 3) No separation was observed during addition of powder or
during hold period.
- 15 4) Smooth incorporation of powder into liquid molten ester
was observed.
- 5) Flakes containing powder were hard. No powder was found
on the surface of the flakes.

20

Example #3 (121-86)

DISPERSION OF ZINC OXIDE (Z-COTE) IN FINSOLV® 137

- In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 25 grams FINSOLV® 137. Melted the flakes at
25 60°C to attain clear liquid on hot water bath. Started
adding zinc oxide powder (25 grams) in five equal portions
over a period of two hours with good mixing and raising the
temperature to 65°C and maintaining the fluidity of the
mixture all the time. Raised the temperature to 70°C and
30 held for one hour with good mixing. Cooled the mixture to
60°C and discharged onto glass pyrex plate. Remarks:
- 1) Addition of zinc oxide powder into liquid ester went very
smoothly, no lumps or clumps.
- 35 2) The mixture was liquid all the time during the addition
of powder and hold period.
- 3) Excellent incorporation of powder into liquid ester. No
separation was observed with one uniform mixture.
- 40 4) Flakes containing powder were hard and brittle.

- 5 5) There was no powder on the surface of the flakes.

Example #4 (121-87)

DISPERSION OF ZINC OXIDE (Z-COTE) IN FINSOLV® 137 &
BEHENYL ALCOHOL

10 In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 22.5 grams FINSOLV® 137 and 5.0 grams Behenyl
Alcohol. Melted the flakes at 55°C to attain clear liquid on
hot water bath. Started adding zinc oxide powder (22.5
grams) in five equal portions over a period of two hours with
15 good mixing and raising the temperature to 65°C and maintain-
ing the fluidity of the mixture all the time. Raised the
temperature to 70°C and held for one hour with good mixing.
Cooled the mixture to 60°C and discharged onto glass pyrex
plate.

20 Remarks:

- 1) Addition of zinc oxide powder into liquid ester went very
smoothly, no lumps or clumps.
- 2) The mixture was liquid and somewhat thinner compared to
25 previous Example 3 during the addition of powder and hold
period.
- 3) Smooth incorporation of powder into liquid ester. No
separation was observed during the addition or hold
30 period; one uniform mixture.
- 4) Flakes containing powder were hard.
- 5) There was no powder found on the surface of the flakes.

35 Example #5 (121-88)

DISPERSION OF ZINC OXIDE (Z-COTE) IN FINSOLV® 116 & CETYL
ALCOHOL

40 In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 22.5 grams FINSOLV® 116 and 5.0 grams cetyl
alcohol. Melted the flakes at 55°C to attain clear liquid on
hot water bath. Started adding zinc oxide powder (22.5
grams) in five equal portions over a period of two hours with
good mixing and raising the temperature to 65°C and maintain-

5 ing the fluidity of the mixture all the time. Raised the temperature to 70°C and held for one hour with good mixing. Cooled the mixture to 60°C and discharged onto glass pyrex plate.

Remarks:

- 10 1) Addition of zinc oxide powder into liquid ester went very smoothly, no lumps or clumps.
- 15 2) The mixture was liquid and somewhat thinner compared to previous Example 2 during the addition of powder and hold period.
- 20 3) Smooth incorporation of powder into liquid ester. No separation was observed during the addition or hold period; one uniform mixture.
- 4) Flakes containing powder were hard.
- 5) There was no powder found on the surface of the flakes.

25

Example #6 (121-89)

DISPERSION OF ZINC OXIDE (Z-COTE) IN FINSOLV® 137 & FINSOLV® TPP

30 In 250 ml glass beaker equipped with thermometer, metallic stirrer added 22.5 grams FINSOLV® 137 and 5.0 grams FINSOLV® TPP. Melted the flakes at 55°C to attain clear liquid on hot water bath. Started adding zinc oxide powder (22.5 grams) in five equal portions over a period of two hours with good

35 mixing and raising the temperature to 65°C and maintaining the fluidity of the mixture all the time. Raised the temperature to 70°C and held for one hour with good mixing. Cooled the mixture to 60°C and discharged onto glass pyrex plate.

40

Remarks:

- 45 1) Addition of zinc oxide powder into liquid ester went very smoothly, no lumps or clumps.
- 2) The mixture was liquid and somewhat thinner compared to previous Example 3 during the addition of powder and hold period.

- 5 3) Smooth incorporation of powder into liquid ester. No
 separation was observed during the addition or hold
 period; one uniform mixture.
- 10 4) Flakes containing powder were hard.
- 5) There was no powder found on the surface of the flakes.

Example #7 (121-90)

DISPERSION OF ZINC OXIDE (Z-COTE HP1) IN FINSOLV® 116

- 15 In 250 ml glass beaker equipped with thermometer, metallic
 stirrer added 25 grams FINSOLV® 116. Melted the flakes at
 55°C to attain clear liquid on hot water bath. Started
 adding zinc oxide powder (25 grams) in five equal portions
 over a period of two hours with good mixing and raising the
20 temperature to 65°C and maintaining the fluidity of the
 mixture all the time. Raised the temperature to 70°C and
 held for one hour with good mixing. Cooled the mixture to
 60°C and discharged onto glass pyrex plate.

Remarks:

- 25 1) Addition of zinc oxide powder into liquid ester went
 very smoothly, no lumps or clumps.
- 2) The mixture was liquid during the addition of powder and
 hold period.
- 30 3) Smooth incorporation of powder into liquid ester. No
 separation was observed during the addition or hold
 period; one uniform, paste-like mixture.
- 35 4) Flakes containing powder were hard and slippery on the
 skin.
- 5) There was no powder found on the surface of the flakes.

40

Example #8 (121-91)

DISPERSION OF ZINC OXIDE (Z-COTE HP1) IN FINSOLV® 137

- 45 In 250 ml glass beaker equipped with thermometer, metallic
 stirrer added 25 grams FINSOLV® 137. Melted the flakes at

- 5 55°C to attain clear liquid on hot water bath. Started
adding zinc oxide powder (25 grams) in five equal portions
over a period of two hours with good mixing and raising the
temperature to 65°C and maintaining the fluidity of the
mixture all the time. Raised the temperature to 70°C and
10 held for one hour with good mixing. Cooled the mixture to
60°C and discharged onto glass pyrex plate. Remarks:
- 1) Addition of zinc oxide powder into liquid ester went very
smoothly, no lumps or clumps. Non-agglomerating and non-
dusting addition process.
 - 15 2) The mixture was liquid during the addition of powder and
hold period.
 - 20 3) Smooth incorporation of powder into liquid ester. No
separation was observed during the addition or hold pe-
riod; one uniform, paste-like mixture.
 - 25 4) Flakes containing powder were hard, brittle and slippery
on the skin.
 - 5) There was no powder found on the surface of the flakes.

Example #9 (121-195)

DISPERSION OF TITANIUM DIOXIDE (X-161) IN FINSOLV® 137

- 30 In 250 ml glass beaker equipped with thermometer, metallic
stirrer added 50 grams FINSOLV® 137. Melted the flakes at
55°C to attain clear liquid on hot water bath. Started
adding titanium dioxide powder (50 grams) in five equal
portions over a period of two hours with good mixing and
raising the temperature to 65°C and maintaining the fluidity
35 of the mixture all the time. Raised the temperature to 70°C
and held for one hour with good mixing. Cooled the mixture
to 60°C and discharged onto glass pyrex plate. Remarks:
- 40 1) Addition of titanium dioxide powder into liquid ester went
very smoothly, no lumps or clumps. Non-agglomerating and
non-dusting addition process.
 - 2) The mixture was liquid during the addition of powder and
hold period.

- 5 3) Smooth incorporation of powder into liquid ester. No
 separation was observed during the addition or hold pe-
 riod; one uniform, paste-like mixture.
- 10 4) Flakes containing powder were hard and brittle.
- 5) There was no powder found on the surface of the flakes.

Example #10 (121-196)

DISPERSION OF TITANIUM DIOXIDE (X-161) IN FINSOLV® 116

- 15 In 250 ml glass beaker equipped with thermometer, metallic
 stirrer added 50 grams FINSOLV® 116. Melted the flakes at
 55°C to attain clear liquid on hot water bath. Started
 adding titanium dioxide powder (50 grams) in five equal
 portions over a period of two hours with good mixing and
20 raising the temperature to 65°C and maintaining the fluidity
 of the mixture all the time. Raised the temperature to 70°C
 and held for one hour with good mixing. Cooled the mixture
 to 60°C and discharged onto glass pyrex plate. Remarks:
- 25 1) Addition of titanium dioxide powder into liquid ester
 went very smoothly, no lumps or clumps. Non-agglomerating
 and non-dusting addition process.
- 2) The mixture was liquid during the addition of powder and
 hold period.
- 30 3) Smooth incorporation of powder into liquid ester. No
 separation was observed during the addition or hold
 period; one uniform, thin paste-like mixture.
- 35 4) Flakes containing powder were hard and brittle and slip
 pery on the skin.
- 5) There was no powder found on the surface of the flakes.

40 Example #11 (121-197)

DISPERSION OF TITANIUM DIOXIDE (M262) IN FINSOLV® 137

 In 250 ml glass beaker equipped with thermometer, metallic
 stirrer added 50 grams FINSOLV® 137. Melted the flakes at
 55°C to attain clear liquid on hot water bath. Started

5 adding titanium dioxide powder (50 grams) in five equal
portions over a period of two hours with good mixing and
raising the temperature to 65°C and maintaining the fluidity
of the mixture all the time. Raised the temperature to 70°C
and held for one hour with good mixing. Cooled the mixture
10 to 60°C and discharged onto glass pyrex plate.

Remarks:

- 1) Addition of titanium dioxide powder into liquid ester went
very smoothly, no lumps or clumps. Non-agglomerating and
non-dusting addition process.
- 15 2) The mixture was liquid during the addition of powder and
hold period.
- 20 3) Smooth incorporation of powder into liquid ester. No
separation was observed during the addition or hold pe
riod; one uniform, paste-like mixture.
- 25 4) Flakes containing powder were hard and brittle, and
slippery on the skin.
- 5) There was no powder found on the surface of the flakes.

PERSONAL CARE PRODUCT FORMULATIONS

30 To further demonstrate the superiority of the delivery
system of the invention, a series of formulations was pre-
pared as described in Example Nos. 12 - 25.

Example #12 (121-106)

35 MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING ZINC OXIDE
(Z-COTE)

INGREDIENTS (INCI)

% BY WT.

40	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
45	B. C12-15 Alkyl Benzoate (FINSOLV® TN)	8.00
	Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
	C. Zinc Oxide (Z-COTE)	8.00

5	D. Propylene Glycol, Diazolidinylurea Methyl Paraben & Propyl Paraben (GERMABEN II)	1.00
		<hr/> 100.00

10 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
- 15 4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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Example #13 (121-110)MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING ZINC OXIDE
(Z-COTE HP1)

25

<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
A. Water	72.75
Hydroxyethyl cellulose (NATROSOL®)	0.25
30 B. C12-15 Alkyl Benzoate (FINSOLV® TN)	8.00
Glyceryl Monostearate	4.00
DEA Oleth-3 Phosphate	3.00
Cyclomethicone (345)	3.00
35 C. Zinc Oxide (Z-COTE HP1)	8.00
D. Propylene Glycol, Diazolidinylurea	1.00
Methyl Paraben & Propyl Paraben (GERMABEN II)	
40	<hr/> 100.00

Procedure:

- 45 1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
- 50 5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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5 Example #14 (121-114)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& ZINC OXIDE (Z-COTE) (OTHER PROCESS):

10	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
15	B. Stearyl Benzoate (FINSOLV® 116)	8.00
	Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
20	C. Zinc Oxide (Z-COTE)	8.00
	D. Propylene Glycol, Diazolidinylurea	1.00
	Methyl Paraben & Propyl Paraben	
	(GERMABEN II)	
25		<u>100.00</u>

Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

Example #15 (121-113)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& ZINC OXIDE (Z-COTE) (INVENTION PROCESS)

	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
45	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
	B. Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
50	Cyclomethicone (345)	3.00
	C. Stearyl Benzoate/Zinc Oxide Flakes	16.00
	(Product of Ex. 2 (121-85))	

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5	D. Propylene Glycol, Diazolidinylurea Methyl Paraben & Propyl Paraben (GERMABEN II)	1.00
		<u>100.00</u>

10 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
- 15 4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

20

Example #16 (121-115)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137,
& ZINC OXIDE (Z-COTE) (OTHER PROCESS):

25

<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
A. Water	72.75
Hydroxyethyl cellulose (NATROSOL®)	0.25
30 B. Behenyl Benzoate (FINSOLV® 137)	8.00
Glyceryl Monostearate	4.00
DEA Oleth-3 Phosphate	3.00
Cyclomethicone (345)	3.00
35 C. Zinc Oxide (Z-COTE)	8.00
D. Propylene Glycol, Diazolidinylurea	1.00
Methyl Paraben & Propyl Paraben (GERMABEN II)	
40	<u>100.00</u>

Procedure:

- 45 1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
- 50 5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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5 Example #17 (121-116)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137
& ZINC OXIDE (Z-COTE) (INVENTION PROCESS)

10	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
15	B. Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
20	C. Behenyl Benzoate/Zinc Oxide Flakes (Product of Ex. 3 (121-86))	16.00
	D. Propylene Glycol, Diazolidinylurea	1.00
	Methyl Paraben & Propyl Paraben (GERMABEN II)	
25		<hr/> 100.00

Procedure:

- | | |
|----|--|
| 30 | 1. Disperse cellulose in water. Heat to 75°C. |
| | 2. Weigh (B) items. Heat to 75°C. |
| | 3. Add (B) to (A) with mixing. |
| | 4. Add (C) in small portions with vigorous stirring over a
period of one hour |
| | 5. Cool to 40°C. Adjust loss of water. |
| 35 | 6. At 40°C add (D). |
| | 7. Homogenize the lotion with hand homogenizer. |

40 Example #18 (121-117)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& ZINC OXIDE (Z-COTE HP1) (OTHER PROCESS):

45	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
	B. Stearyl Benzoate (FINSOLV® 116)	8.00
	Glyceryl Monostearate	4.00
50	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
	C. Zinc Oxide (Z-COTE HP1)	8.00
55	D. Propylene Glycol, Diazolidinylurea	1.00

5 Methyl Paraben & Propyl Paraben
 (GERMABEN II)

100.00

10 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a
15 period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

20 Example #19 (121-118)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& ZINC OXIDE (Z-COTE HP1) (INVENTION PROCESS)

25 INGREDIENTS (INCI)

% BY WT.

A. Water	72.75
Hydroxyethyl cellulose (NATROSOL®)	0.25
B. Glyceryl Monostearate	4.00
DEA Oleth-3 Phosphate	3.00
Cyclomethicone (345)	3.00
C. Stearyl Benzoate/Zinc Oxide Flakes	16.00
(Product of Ex. 7 (121-90))	
D. Propylene Glycol, Diazolidinylurea	1.00
Methyl Paraben & Propyl Paraben	
(GERMABEN II)	

100.00

40 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
45 2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a
period of one hour
5. Cool to 40°C. Adjust loss of water.
50 6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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5 Example #20 (121-194)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137
& ZINC OXIDE (Z-COTE HP1) (OTHER PROCESS):

10	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
15	B. Behenyl Benzoate (FINSOLV® 137)	8.00
	Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
20	C. Zinc Oxide (Z-COTE) (HP1)	8.00
	D. Propylene Glycol, Diazolidinylurea	1.00
	Methyl Paraben & Propyl Paraben	
	(GERMABEN II)	
25		<hr/> 100.00

Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

Example #21 (121-193)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137
& ZINC OXIDE (Z-COTE HP1) (INVENTION PROCESS)

40	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
45	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
	B. Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
50	C. Behenyl Benzoate/Zinc Oxide Flakes	16.00
	(Product of Ex. 8 (121-91))	

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5	D. Propylene Glycol, Diazolidinylurea Methyl Paraben & Propyl Paraben (GERMABEN II)	1.00
		<hr/> 100.00

10 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
- 15 4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

20 Example #22 (122-02)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& TITANIUM DIOXIDE (X-161) (OTHER PROCESS):

25	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
30	B. Stearyl Benzoate (FINSOLV® 116)	8.00
	Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
35	C. Titanium Dioxide	8.00
	D. Propylene Glycol, Diazolidinylurea	1.00
	Methyl Paraben & Propyl Paraben (GERMABEN II)	
40		<hr/> 100.00

Procedure:

- 45 1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
- 50 5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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5 Example #23 (121-040)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 116
& TITANIUM DIOXIDE (X-161) (INVENTION PROCESS)

10	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
15	B. Glyceryl Monostearate	4.00
	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
20	C. Stearyl Benzoate/Zinc Oxide Flakes	16.00
	(Product of Ex. 10 (121-196))	
	D. Propylene Glycol, Diazolidinylurea	1.00
	Methyl Paraben & Propyl Paraben	
	(GERMABEN II)	
25		<hr/> 100.00

Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

Example #24 (122-01)

40 MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137
& TITANIUM DIOXIDE (X-161) (OTHER PROCESS):

	<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
45	A. Water	72.75
	Hydroxyethyl cellulose (NATROSOL®)	0.25
	B. Behenyl Benzoate (FINSOLV® 137)	8.00
	Glyceryl Monostearate	4.00
50	DEA Oleth-3 Phosphate	3.00
	Cyclomethicone (345)	3.00
	C. Titanium Dioxide (x-161)	8.00

55

5	D. Propylene Glycol, Diazolidinylurea Methyl Paraben & Propyl Paraben (GERMABEN II)	1.00
		<u>100.00</u>

10 Procedure:

1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
- 15 4. Add (C) in small portions with vigorous stirring over a period of one hour
5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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Example #25 (122-03)

MOISTURIZING SUNSCREEN LOTION FORMULA CONTAINING FINSOLV® 137
& TITANIUM DIOXIDE (X-161) (INVENTION PROCESS)

25

<u>INGREDIENTS (INCI)</u>	<u>% BY WT.</u>
A. Water	72.75
Hydroxyethyl cellulose (NATROSOL®)	0.25
30 B. Glyceryl Monostearate	4.00
DEA Oleth-3 Phosphate	3.00
Cyclomethicone (345)	3.00
35 C. Behenyl Benzoate/Titanium Dioxide Flakes (Product of Ex. 9 (121-195))	16.00
D. Propylene Glycol, Diazolidinylurea	1.00
Methyl Paraben & Propyl Paraben (GERMABEN II)	
40	<u>100.00</u>

Procedure:

- 45 1. Disperse cellulose in water. Heat to 75°C.
2. Weigh (B) items. Heat to 75°C.
3. Add (B) to (A) with mixing.
4. Add (C) in small portions with vigorous stirring over a period of one hour
- 50 5. Cool to 40°C. Adjust loss of water.
6. At 40°C add (D).
7. Homogenize the lotion with hand homogenizer.

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5 **Remarks on Examples of Invention Process**
 (Example Nos. 15, 17, 19, 21, 23, and 25)

 The process of the invention comprises pre-incorporating
physical sunscreens, e.g., titanium dioxide or zinc oxide,
into the specified benzoate esters before dispersing into the
10 combined phases A & B. Thus, a blend of physical sunscreen
with benzoate esters is pre-formed in the form of solid
chunks or flakes and used to prepare sunscreen lotions. The
following observations were made:

15 1. The addition of micronized zinc oxide powder or tita-
nium dioxide powder pre-incorporated in stearyl and behenyl
benzoates in solid form, preferably in the form of chunks or
flakes, into combined portion of A & B was very, very easy.
As each portion of the flakes melted in the combined portion
20 A & B it became a part of the system. No dust or any kind of
dusting problem whatsoever occurred. In particular, in
Example Nos. 7 & 8, where dimethicone was present, it gives a
smooth lotion at the end and helps during the process of
making cosmetic lotions due to homogeneity and ease of dis-
25 persion.

 2. In every Example using the invention process, there was
not a single case of agglomeration. The process is non-
agglomerating.

30 3. In each example of the use of the invention process
(Example Nos. 15, 17, 19, 21, 23, and 25), a uniform lotion
was obtained. As soon as the pre-incorporated blend in its
solid flake or chunk form was added to the combined phase A &
35 B, the sunscreen/ester blend went into the system without any
difficulty as the individual portions of our invention prod-
uct (Phase C) were being added progressively. There was no
separation of any particles nor was there any stickiness on

5 the sides of the vessel. The invention process-based blends
give uniform lotions.

4. As soon as the chunks or flakes went into the combined
mixture A & B, the chunks or flakes of the pre-blended prod-
10 ucts of this invention melted fast, and did not find any
powder around or underneath the bottom of the mixer.

5. The incorporation of physical sunscreen powder in the
form of solid chunks or flakes of FINSOLV® 116 and FINSOLV®
15 137 (stearyl & behenyl benzoates) into water and oil phase
was very easy. As the flakes melted, it becomes part of the
system with the sunscreen in it.

6. During homogenizing of the lotion with a hand homoge-
20 nizer, did not feel the sound of the particles, i.e., gritti-
ness of the particles. Before going through the homogenizer,
the particles are well mixed and become part of the lotion.

7. After homogenizing, the spreadability of the lotion was
25 excellent compared to the Examples using the other, known
process. It was a very uniform lotion.

8. Lotions of Example Nos. 15, 17, 19, 21, 23, and 25) did
not show whitening effect. No disruption of the particle
30 distribution, i.e., there were no larger chunks in the lo-
tion, it was all finely dispersed.

Remarks on Examples of Other, Prior Art Process
(Example Nos. 12-14, 16, 18, 20, 22, and 24)

35 The other, known process comprises adding physical
sunscreens, i.e., titanium dioxide or zinc oxide, separately
into the combined mixture of phases A & B. Benzoate esters
and physical sunscreens are added separately to make the

5 sunscreen lotions. The following observations were made:

1. Addition of micronized zinc oxide powder or titanium
dioxide powder into combined portion of phases A & B was very
difficult. Powder created lots of dust during addition.
10 Even the presence of dimethicone in zinc oxide powder, and
stearic acid and alumina in titanium dioxide, did not resolve
the dusting problem.

2. In every Example of other process, i.e., Example Nos.
15 12-14, 16, 18, 20, 22, and 24, found formation of agglomera-
tion of the particles.

3. In all cases, it was very difficult to make a uniform
lotion. Powder tries to scatter in every direction with
20 mixing. Decreasing the speed of the mixer created lumps
around the mixer.

4. Water phase with physical sunscreen powder tries to
stick around the mixer and beneath the mixer which creates a
25 disproportionate amount of the sunscreen in the lotion.

5. Incorporation of physical sunscreen powder into oil and
water phases was very cumbersome and tough.

30 6. When homogenizing the lotion with hand homogenizer,
there was a distinct sound of the particles, i.e., grittiness
of the particles, which was squeezing through the nozzle of
the homogenizer.

35 7. After homogenizing, the spreadability of the lotion was
very poor. The lotions were thick and non-uniform, i.e.,
heterogeneous.

- 5 8. Lotions showed whitening effect due to the uneven
distribution of the particles in the system.

10 It will be understood that the embodiments described herein
are merely exemplary and that a person skilled in the art may
make many variations and modifications without departing from
the spirit and scope of the invention. All such variations
and modifications are intended to be included in the scope of
the invention as described herein.

5 We claim:

1. A method of preparing a solid dispersion of physical, inorganic sunscreens for use in sunscreen preparations comprising the steps of:

- 10 a. melting a solid dispersion vehicle;
b. adding one or more inorganic sunscreen agents to said melted dispersion vehicle;
c. stirring and heating the mixture to a temperature and for a time sufficient to disperse said sunscreen agent in
15 said dispersion vehicle;
d. after the sunscreen agent is dispersed in said dispersion vehicle, cooling the mixture while stirring to thereby produce a solid dispersion of said one or more inorganic sunscreen agents for use in sunscreen preparations.

20 2. The method of claim 1 wherein said solid dispersion vehicle is selected from the group consisting of esters of benzoic acid and alcohols having from 16 to 22 carbons, and solid fatty alcohols selected from the group consisting of
25 Cetyl Alcohol, Stearyl Alcohol, Cetearyl Alcohol, Behenyl Alcohol, Arachidyl Alcohol and higher alcohols having 22+ carbon atoms.

30 3. The method of claim 2 wherein said solid dispersion vehicle is preferably selected from the group consisting of stearyl benzoate, behenyl benzoate and arachidyl benzoate.

35 4. The method of claim 1 wherein said inorganic sunscreen agent is selected from the group consisting of micronized titanium dioxide and zinc oxide.

5. The method of claim 1 wherein the temperature to which the mixture is heated is above 60°C.

5 6. The method of claim 1 wherein between about 10% and 60% of one or more inorganic sunscreen agents is added to between about 40% and 90% of said melted dispersion vehicle.

10 7. The method of claim 1 wherein after step b., adding between 1 to 20% by weight based on the weight of the inorganic sunscreen of an emollient selected from the group consisting of C₁₂₋₁₅ Alkyl Benzoate (FINSOLV® TN from Finetex Inc.) Octyl Dodecyl Benzoate (FINSOLV® BOD from Finetex Inc.), PPG-15 Stearyl Ether Benzoate (FINSOLV® P from Finetex Inc.) and Dipropylene Glycol Dibenzate (FINSOLV® PG-22 from Finetex Inc.).

20 8. The method of claim 1 wherein said solid dispersion while cooling is subjected to flaking.

20 9. The method of claim 1 further comprising, after step d., discharging said solid dispersion on a cooled surface.

25 10. A sunscreen preparation comprising a cosmetically acceptable carrier and an effective amount of a solid dispersion of said one or more inorganic sunscreen agents according to claim 1 as the active sunscreen ingredient.

30 11. The sunscreen preparation of claim 10 wherein said solid dispersion is from about 5% to about 50% by weight of said sunscreen preparation.

35 12. A method of protecting human skin or hair from ultraviolet radiation comprising applying to the skin or hair an effective quantity of a sunscreen preparation according to claim 10.

13. A homogeneous, storage-stable, solid dispersion of micronized inorganic sunscreen comprising a solid dispersion

5 vehicle selected from the group consisting of stearyl benzoate, behenyl benzoate and arachidyl benzoate, and one or more inorganic sunscreen agents selected from the group consisting of micronized titanium dioxide and zinc oxide.

10 14. The solid dispersion of claim 13 comprising between about 40% and 90% of said solid dispersion vehicle and between about 10% and 60% of said one or more inorganic sunscreen agents.

15 15. The solid dispersion of claim 13 preferably comprising 50% of said solid dispersion vehicle and 50% of said one or more inorganic sunscreen agents.

20 16. A homogeneous, storage-stable, solid dispersion of one or more inorganic sunscreen agents selected from the group consisting of micronized titanium dioxide and micronized zinc oxide and a solid dispersion vehicle selected from the group consisting of stearyl benzoate, behenyl benzoate and arachidyl benzoate, wherein said solid dispersion is prepared
25 by melting said solid dispersion vehicle, adding said one or more inorganic sunscreen agents, stirring and heating the mixture to a temperature and for a time sufficient to disperse said sunscreen agent in said dispersion vehicle, and after said sunscreen agent is dispersed in said dispersion
30 vehicle, cooling said mixture while stirring to thereby produce a solid dispersion of inorganic sunscreen, and discharging said solid dispersion on a cooled surface.

35 17. The solid dispersion of claim 16 comprising between about 10% and 60% of one or more of said inorganic sunscreen agents blended into between about 40% and 90% of said dispersion vehicle.

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18. A sunscreen preparation comprising a cosmetically acceptable carrier and from about 5% to about 50% of the solid dispersion of one or more inorganic sunscreen agents of claim 16.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US01/08330

A. CLASSIFICATION OF SUBJECT MATTER IPC(7) : A61K 7/42 US CL : 424/401, 59, 70.1, 70.9 According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) U.S. : 424/401, 59, 70.1, 70.9 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) KOSMET, USPATFULL, HCAPLUS		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,917,882 A (STROGRIDGE) 17 April 1990, see whole document.	1-3
Y	US 4,323,693 A (SCALA, Jr.) 06 April 1982, see whole document.	1, 4-18
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
* *A* *E* *L* *O* *P*	Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance earlier document published on or after the international filing date document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) document referring to an oral disclosure, use, exhibition or other means document published prior to the international filing date but later than the priority date claimed	*T* *X* *Y* *A* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art document member of the same patent family
Date of the actual completion of the international search 30 APRIL 2001		Date of mailing of the international search report 26 JUL 2001
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